AD 728175

THE STUDY OF THE INTERACTION OF INTENSE PICOSECOND LIGHT PULSE WITH MATERIALS

A QUARTERLY TECHNICAL REPORT

SUBMITTED TO

THE U.S. ARMY RESEARCH OFFICE

PERIOD

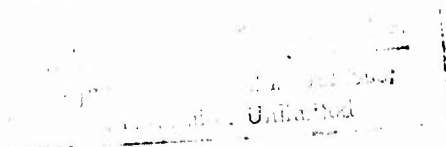June 19, 1970 to September 18, 1970

REPORTED BY

CHI H. LEE

TECHNICAL
RESEARCH
REPORT

DEPARTMENT OF
ELECTRICAL ENGINEERING

UNIVERSITY OF MARYLAND

COLLEGE PARK, MARYLAND 20742

17

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified;

1. ORIGINATING ACTIVITY (Corporate author)	2a. REPORT SECURITY CLASSIFICATION
Department of Electrical Engineering University of Maryland College Park, Maryland 20742	Unclassified
	2b. GROUP

3. REPORT TITLE

The Study of the Interaction of Intense Picosecond Light Pulses with Material (The Observation of Multiphoton-induced conductivity with Mode-locked Lasers.

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)

A quarterly technical report, June 19, 1970 to Sept. 18, 1970

5. AUTHOR(S) (First name, middle initial, last name)

Chi H. Lee

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
Oct. 15, 1970	14	10

8a. CONTRACT OR GRANT NO.	9a. ORIGINATOR'S REPORT NUMBER(S)
DA-ARO-D-31-124-70-G50	
b. PROJECT NO. ARPA Order No. 675, Am. 7	
c. Program Code No. 9E20	9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)
d.	

10. DISTRIBUTION STATEMENT

Reproduction in whole or in part is permitted for any purpose of the United States Government.

11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY
	ARPA and U. S. Army Research Office

13. ABSTRACT

This report covers work under contract DA-ARO-D-31-124-70-G50 for the period June 19 to Sept. 18, 1970. Topics discussed include photoconductivity effect in GaAs induced by picosecond light pulses from a mode-locked Nd: glass laser. In addition to the two-photon absorption, we found that the observed effect also involved stimulated hole scattering and three-photon absorption.

14 KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Laser						
Picosecond light pulses						
Two-photon absorption						
Photoconductivity						
Mode-locked laser						

Quartely Technical Report

For

Period June 19, 1970 to September 18, 1970

Submitted to the U.S. Army Research Office

ARPA Order Number	675, Am. 7
Program Code Number:	9E20
Name of Grantee:.	University of Maryland
Effective Date of Grant:	December 18, 1969
Grant Expiration Date:	December 19, 1970
Amount of Grant:	$30,000
Principal Investigator and Phone Number	Dr. Chi H. Lee (301) 454-2443
Grant Number:	DA-ARO-D-31-124-70-G50
Project Scientist or Engineer	None
Short Title of Work:	"The Study of the Interaction of Intense Picosecond Light Pulse with Materials"

Reported by: _____

Dr. Chi H. Lee
Assistant Professor

The Observation of Multiphoton-induced Conductivity

with Mode-locked Lasers

The photoconductivity of semi-insulating GaAs (Cr doped) Crystal was investigated with mode-locked Nd-glass laser pulses. The log-log plot of photoconductivity in milli mhos versus relative intensity is shown as curve B in Figure 1. The theoretical value of the two photon conductivity can be calculated using the formula[1]

$$\Delta G = q(\mu_e + \mu_r) \frac{\beta L I_o^2}{1 + \beta L I_o} \times \frac{10^{+6} x t_i}{2\hbar w \times 1.6 \times 10^{-19}}$$

$$= (\mu_e + \mu_r) \frac{\beta L I_o^2}{1 + \beta L I_o} \times \frac{t_i}{2\hbar w} \times 10^6 \text{ mhos.}$$

where I_o is in Mw/cm^2

    $\hbar w$ - photon energy = 1.17 ev

 β - two photon absorption coefficient in cm/Mw = $5.6 \frac{cm}{Mw}$

    L - thickness of crystal = .033 cms.

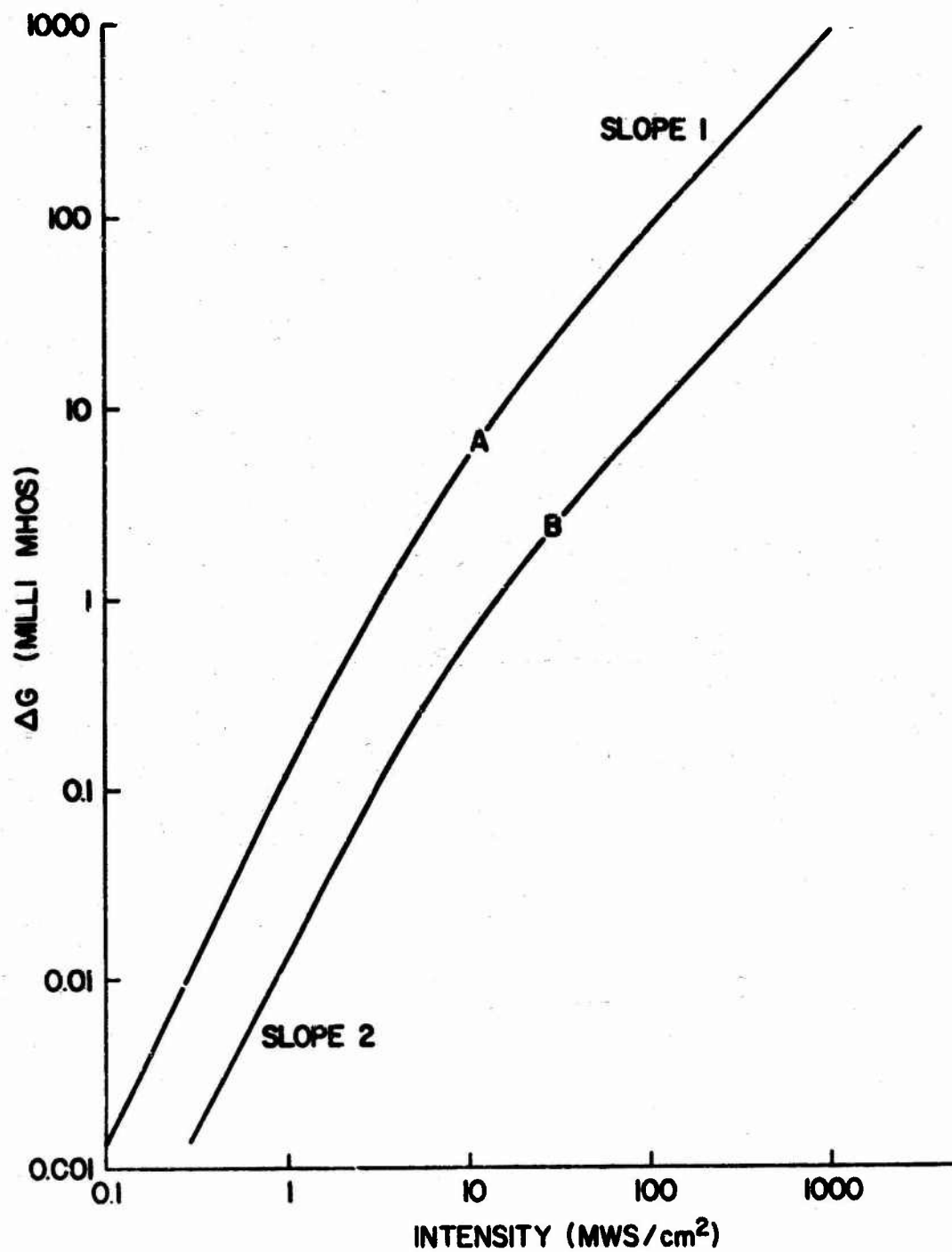$\mu_e + \mu_r$ = 230 cm^2/v-sec.

    t_i = pulse width (1 p sec.).

Since the photoconductivity is induced by mode locked pulses, the short time width of the pulse (of the order of pico second) enters into the calculation of ΔG instead of life time. This was confirmed by the observation of structure of the p. c. pulses in the 519 oscilloscope. The calculated value of two photon conductivity is shown in Figure 2 against absolute intensity. By comparing Figure 1 and Figure 2, we conclude that at lower

Fig. 1. The experimental observed photo-conductivity change verse incident laser intensity.
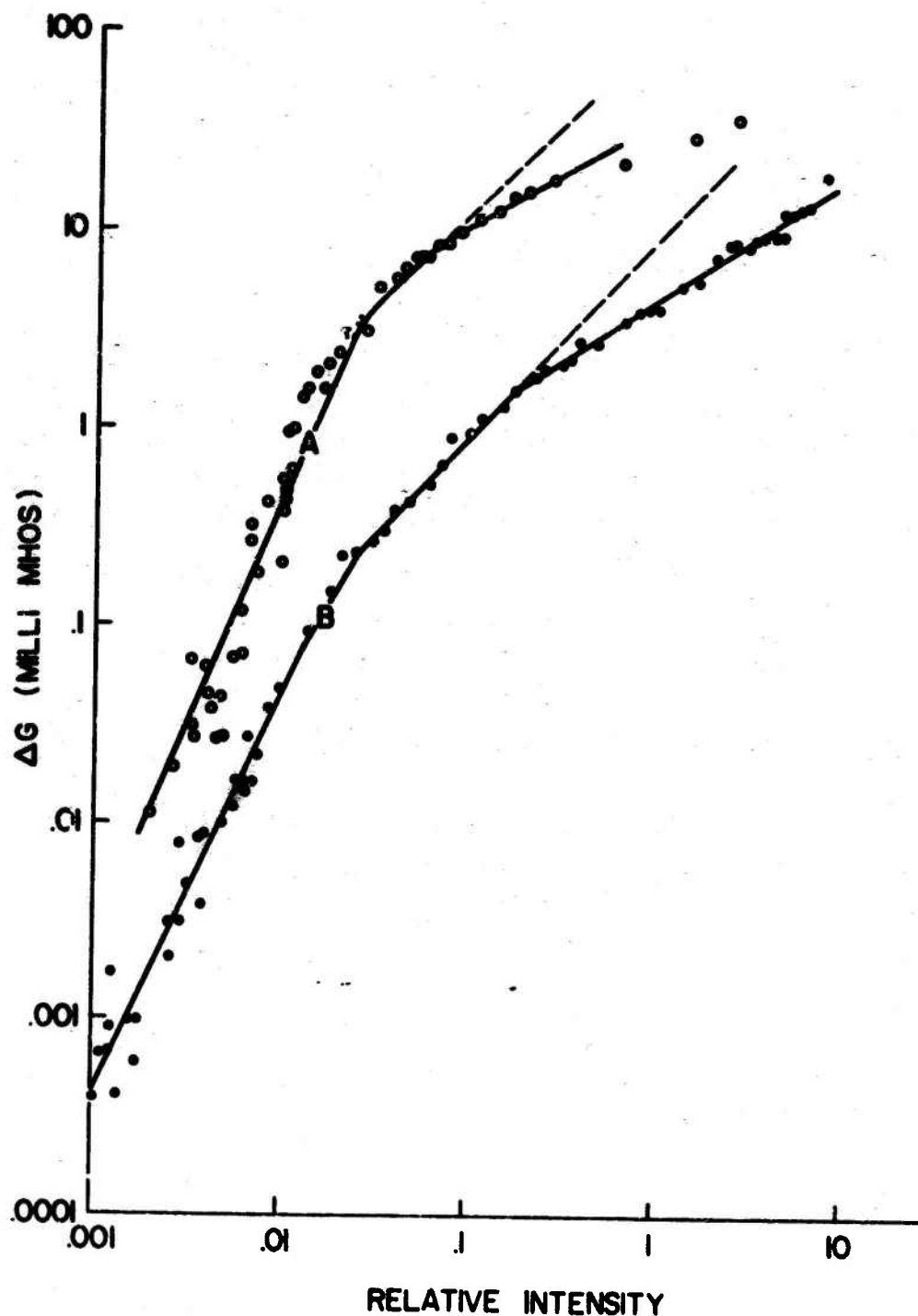
CALCULATED TWO PHOTON
CONDUCTIVITY

A   n−TYPE O_2 DOPED GaAs
(.028 cm THICK, $\mu_e + \mu_r = 7000$ cm^2/v−sec)

B   Cr−DOPED SEMI INSULATING GaAs
(.033 cm THICK, $\mu_e + \mu_r \approx 235$ cm^2/v−sec)

LIFE TIME $\tau \gg t_i$ (pulse width) $\approx$ l p sec

Fig. 2.   Calculated two photon conductivity as a function of incident laser
intensity.

intensities the shape of the experimental curve is more or less the same as that of the theoretical one. A slope of 2 changing to unity slope at higher intensities is observed indicating the two photon excitation of conductivity. If the conductivity is due to two photon excitation alone, we should observe a slope of 1 at very high intensities with increase of intensity. At very high intensities, a nonlinearity is introduced as shown in Fig. 3. The observed conductivity is less than the ideal extrapolated curve of slope unity.

The nonlinearity is definitely not due to the surface recombination since the excitation is due to short pulse whose width is much less than the recombination times.

Since conductivity is directly proportional to mobility, it was first suspected that mobility [2, 3] may decrease at high excitation levels because of electron-hole scattering. This decrease in mobility at high excitation levels in $S_i(4.2^{\circ}K)$ was observed by A. A. Patrin et al. [3] The following discussion rules out this possibility in the present experiment.

At room temperature, the mobility in GaAs is determined by screened polar scattering and charged impurity scattering. [4, 5] Normally at room temperature, the polar scattering dominates. The room temp. mobility $(\mu_e + \mu_r) = 230 \text{ cm}^2/\text{v-sec}$ was estimated by observing the ratio of the conductivities of two different samples (one high resistivity cromium doped and the other one n type O_2 doped low resistivity crystal whose mobility is known). Since the two photon conductivity is in dependent of β in the region of slope 1, the ratio of the conductivities in this region gave a value of
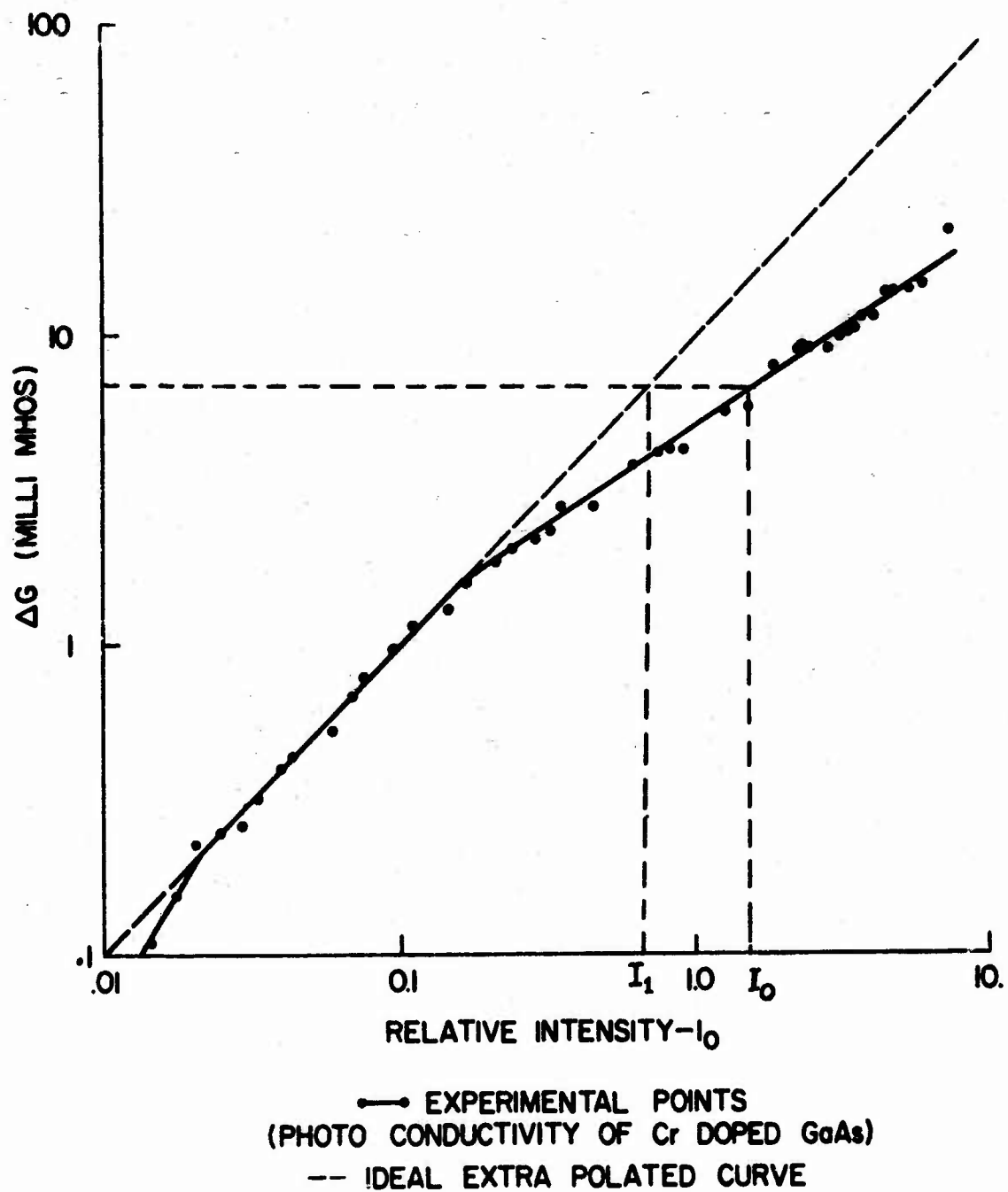
Fig. 3. Observed change of photoconductivity in Cr-doped GaAs crystal for relative high intensity of the laser beam.

6

230 cm^2/v-sec for the present crystal. This rather low value of mobility had been observed for Cr doped crystals by Cronin and Haisty.[6] When the crystal is excited, the density of non equilibrium charge carriers increases. The contribution of mobility due to electron-hole scattering was calculated using the Brooks-Herring formula[7]

$$\mu_{e-h} = \frac{2^{7/2} \, \epsilon^2 (kT)^{3/2} \, (m_e + m_h)^{1/2}}{\pi^{3/2} \, e^3 (m_e m_h)^{1/2} (n_e n_h)^{1/2} \left[\ln(1+B) - \frac{B}{1+B} \right]}$$

where $B = \dfrac{6\epsilon (kT)^2 \, m_e m_h}{\pi \hbar^2 e^2 (m_e + m_h)(n_e n_h)^{1/2}}$

ϵ = dielectric constant.

m_e = electron effective mass.

m_h = hole effective mass.

$n_e = n_h$ = no. of electron hole pairs.

$\epsilon = 11.8$, $m_e = 0.074$ m

$\qquad m_h = m^*_{\theta_1} = 0.68m$ [8],

at $T = 300^\circ K$, the value of μ_{e-h} against the density of electron-hole pairs is calculated. Even at 10^{18}/cm^3 of pair concentration, the mobility is of the order of 10^4 and this does not affect the mobility (low value 230 cm^2/v-sec) at very high intensities of illumination. Thus the observed non-linearity is not due to increase of mobility due to electron-hole scattering.

The second possilbility may be due to the free electron absorption. The free electron absorption[8] in GaAs for a concentration (electron) of

$10^{17}/\text{cm}^3$ is approximately $.01 \text{ cm}^{-1}$ at 1μ wavelength. So the free carrier absorption is low and the observed nonlinearity is not due to this process.

The most likely possibility is that due to free hole absorption (or intra band absorption). The intraband absorption in GaAs with various p-type doping densities had been extensively studied by Brannstein, [9] Brannstein and Kane. [10] The hole absorption coefficient at 1μ for the crystal doped with $10^{17}/\text{cm}^3$ is about 3 cm^{-1}. This absorption coefficient is due to the transitions between the light hole, heavy hole and split-off valence bands. The band structure of GaAs is shown in Figure 4. In the present case, at higher intensities of light, the nonequilibrium charge carriers (electron-hole pairs) are produced proportional to the light intensity and the associated stimulated absorption by the nonequilibrium holes should also increase linearly with the light intensity. Referring to Figure 3., from the nonlinear region, the absorption coefficient due to intraband absorption was calculated as follows. For a particular value of ΔG, the intensities $(I_1 + I_0)$ at the ideal slope 1 curve and experimental photoconductivity curve are determined.

$$I_1 = I_0 e^{-\alpha L} .$$

$$\alpha L = \ln \left(\frac{I_0}{I_1} \right) .$$

The carrier density is proportional to I_1. A log-log plot of αL versus relative intensity I_1 is shown in Figure 5. Again at lower intensities, αL increases linearly with light intensity thereby showing that the
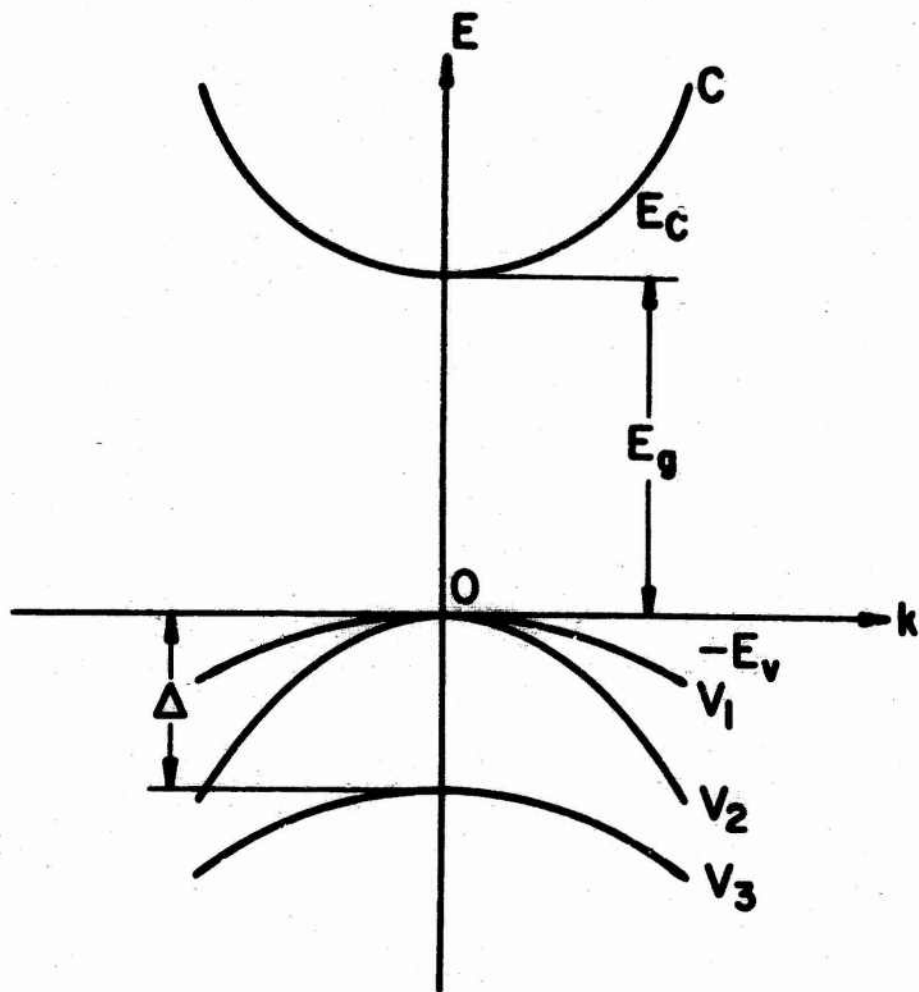
BAND · STRUCTURE OF Ga As

C - conduction band ; $V_{1,2,3}$ - valence bands.

E_g - forbidden gap , Δ - split off band width.

Energy measured w.r.t. the top of valence bands V_1 and V_2.

$E_g = 1.41$ ev , $\Delta = 0.33$ ev (at 300 °k) ;

Effective masses m_c^* , $m_{V_1}^*$, $m_{V_2}^*$, $m_{V_3}^*$ = 0.72 m_0 ,

0.68 m_0 , 0.085 m_0 , 0.25 m_0 ; m_0 - electron rest mass.

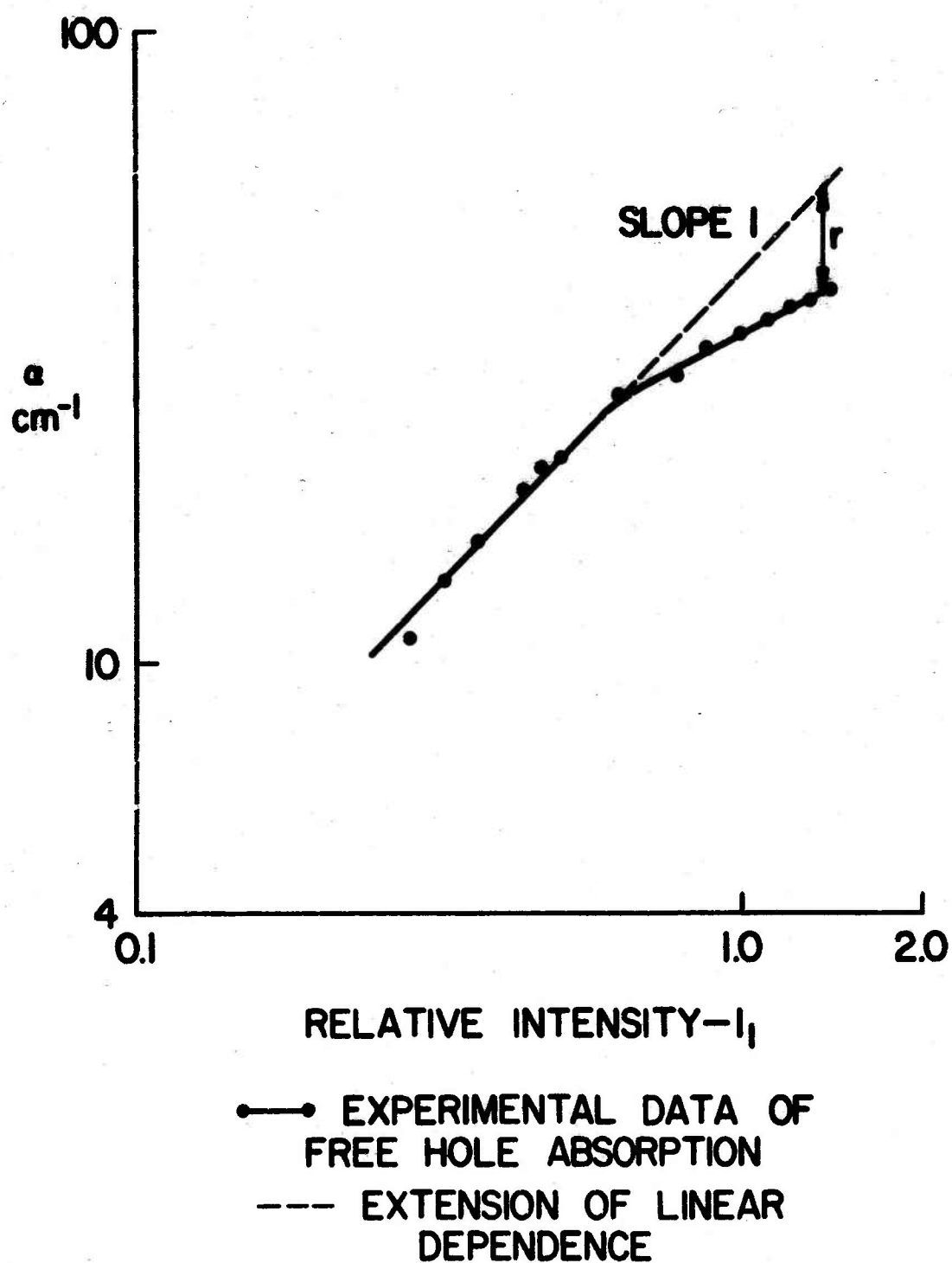Fig. 4. Band structure of GaAs.

Fig. 5. Experimental data of free hole absorption in GaAs under laser excitation.

nonlinearity in the two photon conductivity is due to stimulated intraband absorption. However at very high light intensities, αL increases rather slowly. This means that part of the light intensity is being utilized in the generation of nonequilibrium carriers (producing conductivity). The difference in absorption between the linear one and sublinear one in the αL versus I_1 graph in Figure 4 (γL) is plotted against relative intensity I_o in a log-log graph in Figure 6. The slope of 2 in Figure 6 suggests that there is generation mechanism whose absorption coefficient increases as the square of the light intensity. Hence we conclude that we have observed the three photon generation of nonequilibrium charge carriers in GaAs at very high intensities of light.

Referring to Figure 6, at about 5 relative intensity, (this corresponds to roughly 30 Gw/cm^2), the free hole absorption reads approximately as 10 cm^{-1}. 3000 Mw/cm^2 corresponds to $\dfrac{5 \times 10^9}{1.6 \times 2.34 \times 10^{-19}} \times 10^{-12} \times \dfrac{1}{.033}$

$\approx 4.5 \times 10^{17}/\text{cm}^3$. This corresponds to Ref. [9], an absorption coefficient of 5 cm^{-1}. Our value comes out to be slightly higher which might be due to the fact that the high resistivity crystal has more compensating impurities.

Referring to Figure 6, for 30 Gw/cm^2, we get the three photon absorption coefficient as 10 cm^{-1}. Using the formula derived by A. I. Bohryskeva et al[11] on three-photon band-band transitions in semiconductors, and applying it to GaAs, (valence band v_1 to conduction band), we get for a photon density of 10^{16} cm^{-3}, $K^{(3)} \approx 2 \times 10^{-5}$ cm^{-1}. For 30 Gw/cm^2, we get a calculated
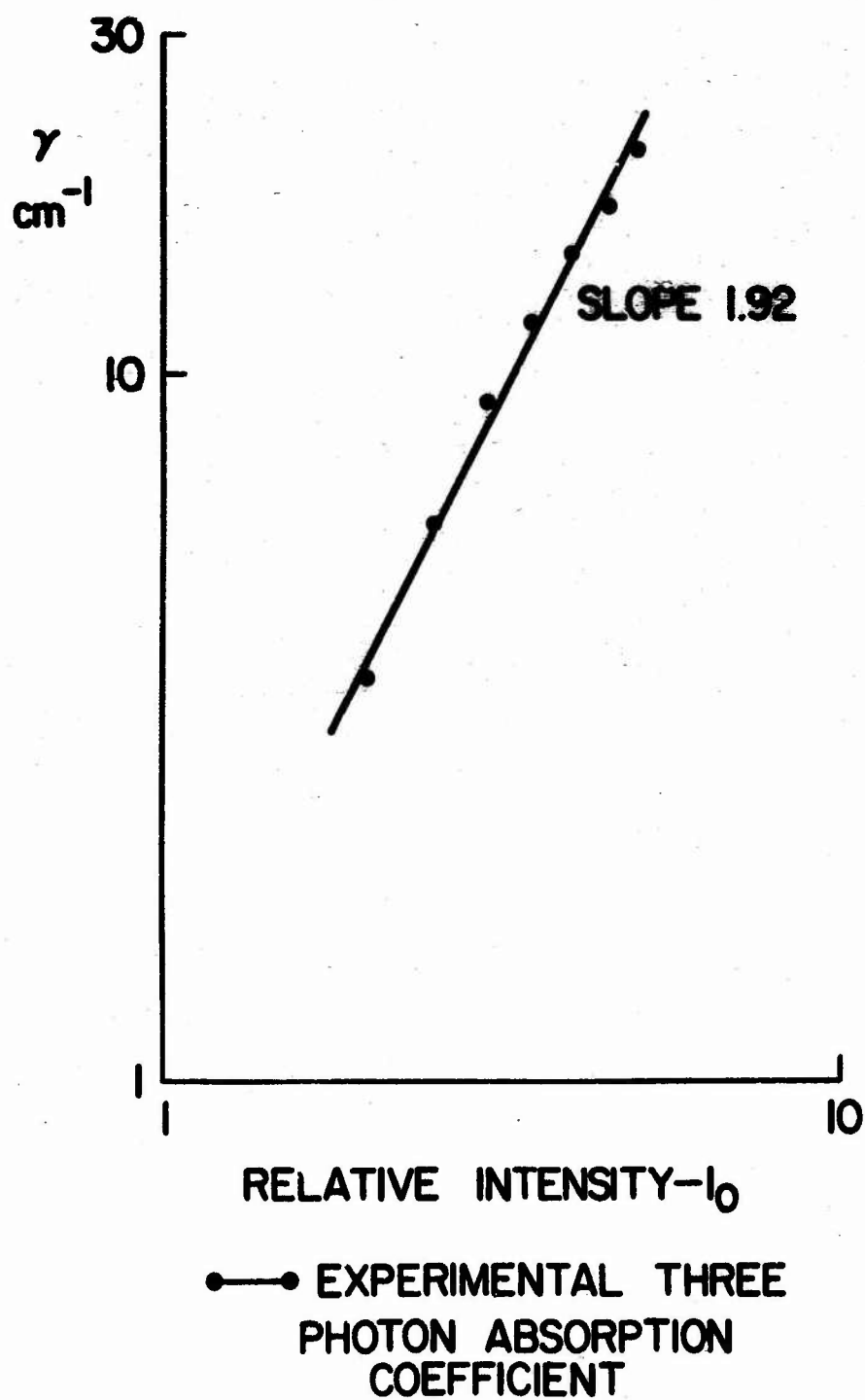
Fig. 6. Experimental three photon absorption coefficient of GaAs.

value 5 cm^{-1} which agrees approximately with the experimentally determined values.

Conclusion

The portion of the ΔG versus I curve agrees with the theoretical two photon conductivity curve at low intensities, the slope at lower intensities being equal to 2 changing to unity at higher intensities. At very high intensities, the curve displays a non linear region which was proved to be due to stimulated intraband absorption of holes in GaAs. The magnitude of the absorption coefficient α varies from 10 to 30 cm^{-1} (αL varies between .35 to .95, L = .033 cm) linearly with intensity. This agrees approximately with the values reported in the references [9], [10]. Towards the end of the high intensity region, absorption which leads to the generation of carriers is observed. This absorption coefficient is found to be proportional to the square of the light intensity indicating the generation of non-equilibrium charge carriers in GaAs due to three photon absorption. The calculated 3 photon absorption coefficient agrees approximately with the experimentally observed value.

References

[1] J. H. Yee, Phys. Rev. vol. 186, No. 3 (1969), 778.

[2] K. D. Glinchuk, et al., Phys. Stat. Sol. 42, K 171 (1970).

[3] A. A. Patrin et al. Soviet Physics-Semiconductors, vol. 3, No. 3, 383, (1969).

[4] H. Ehrenreich, Phys. Rev. 120, (1960) 1951.

[5] Isamu Akasaki and Tohru Hara, J. Phys. Soc. Japan 20, (1965) 2292.

[6] G. R. Cronin and R. W. Haisty, J. Electrochem. Soc. 111, 874 (1964).

[7] F. J. Morin and J. P. Maita, Phy. Rev., 94 1525 (1954).

[8] T. S. Moss and T. D. F. Hawkins, vol. 1, pp. 111-115, Infrared Physics, 1961.

[9] R. Brannstein, J. Phys. Chem. Solids, 8 (1959) 280.

[10] R. Brannstein and E. O. Kane, J. Phy. Chem. Solids 23 (1962) 1423.

14

Figure Captions

Fig. 1.   The experimental observed photo-conductivity change verse incident laser intensity.

Fig. 2.   Calculated two photon conductivity as a function of incident laser intensity.

Fig. 3.   Observed change of photoconductivity in Cr-doped GaAs crystal for relative high intensity of the laser beam.

Fig. 4.   Band structure of GaAs.

Fig. 5.   Experimental data of free hole absorption in GaAs under laser excitation.

Fig. 6.   Experimental three photon absorption coefficient of GaAs.